

In the Specification

Kindly replace paragraph [0001] of the Specification as published with the following:

~~The present invention~~ This disclosure relates to a stainless steel for a proton-exchange membrane fuel cell (or solid-polymer fuel cell) separator having high durability and a low contact resistance and to a proton-exchange membrane fuel cell using the same.

Kindly replace paragraph [0020] of the Specification as published with the following:

In addition, attempts have been made such that stainless steels not subjected to surface treatment are used as they are for separators. For example, Japanese Unexamined Patent Application Publications No. 2000-239806 and No. 2000-294256 each discloses a separator-use ferritic stainless steel formed such that Cu and Ni are positively added, impurity elements such as S, P, and N are then reduced, and $C + N \leq 0.03$ mass% and $10.5 \text{ mass \%} \leq Cr + 3 \times Mo \leq 43$ mass% are satisfied. In addition, Japanese Unexamined Patent Application Publications No. 2000-265248 and No. 2000-294256 each discloses a separator-use ferritic stainless steel formed such that Cu and Ni are restricted to 0.2 mass % or less thereby to inhibit dissolution of metallic ions, impurity elements such as S, P, and N are then reduced, and $C + N \leq 0.03$ mass % and $10.5 \text{ mass \%} \leq Cr + 3 \times Mo \leq 43$ mass % are satisfied.

Kindly replace paragraph [0024] of the Specification as published with the following:

~~Japanese Unexamined Patent Application Publication No. 2003-223904 discloses an invention developed by the present Applicant. The invention even incorporates results of researches research and investigations investigation on of the influence of surface roughness of the surface of stainless steel. In this case, the stainless steel surface roughness is set as: Ra: 0.01 μm to 1 μm ; and Ry: 0.01 to 20 μm . However, the contact resistance was found insufficient from the viewpoint of securing higher cell output.~~

Kindly replace paragraph [0025] of the Specification as published with the following:

DISCLOSURE OF INVENTION

~~In view of the above-described problems with the conventional techniques, the object of the present invention is it could be helpful to provide a stainless steel for a proton-exchange~~

membrane fuel cell separator that has a high corrosion resistance and a low contact resistance (i.e., high electrical conductivity) and to provide a proton-exchange membrane fuel cell using the same.

Kindly replace paragraph [0026] of the Specification as published with the following:

More specifically, ~~the object of the present invention is it could be helpful~~ to provide a stainless steel for a proton-exchange membrane fuel cell separator and a proton-exchange membrane fuel cell using the same, wherein the proton-exchange membrane fuel cell separator is provided by specifying not only composition of stainless steel being as base material, but also composition of a passive film existing on the surface thereof, thereby to have a low contact resistance, a high electric power generation efficiency, and a high corrosion resistance of the stainless steel itself even without being subjected to a surface treatment.

Kindly replace paragraph [0027] of the Specification as published with the following:

Summary

~~In order to achieve the object described above, the present invention has features described hereunder.~~

Kindly replace paragraph [0028] of the Specification as published with the following:

~~In one aspect, the present invention provides~~ We thus provide a stainless steel for a proton-exchange membrane fuel cell separator, having a composition comprising 0.03 mass % or less of C, 16-45 mass % of Cr, 0.03 mass % or less of N, 0.1-5.0 mass % of Mo, wherein a total of the C content and the N content satisfies 0.03 mass % or less; a balance portion is comprised of Fe and unavoidable impurities; with respect to Al, Cr, and Fe contained in a passive film on a surface of the stainless steel an atomic ratio of Cr/Fe is 1 or greater; and an atomic ratio of Al/(Cr+Fe) is less than 0.10.

Kindly replace paragraph [0029] of the Specification as published with the following:

~~In addition, the present invention provides~~ we provide a stainless steel for a proton-exchange membrane fuel cell separator, having a composition comprising 0.03 mass % or less of C, 0.03 mass % or less of N, 20-45 mass % of Cr, and 0.1-5.0 mass % of Mo, wherein a total of

the C content and the N content satisfies 0.03 mass % or less; a balance portion is comprised of Fe and unavoidable impurities; with respect to Al, Cr, and Fe contained in a passive film on a surface of the stainless steel an atomic ratio of Cr/Fe is 1 or greater, and an atomic ratio of Al/(Cr+Fe) is less than 0.05.

Kindly replace paragraph [0030] of the Specification as published with the following:

~~In accordance with the above described aspect, the present invention~~ We further provides provide a stainless steel for a proton-exchange membrane fuel cell separator wherein in addition to the composition, the stainless steel comprises at least one selected from a group of items (1) - (4):

- (1) Si: 1.0 mass % or less;
- (2) Mn: 1.0 mass % or less;
- (3) Al: 0.001-0.2 mass % or less; and
- (4) Ti or Nb: 0.01-0.5 mass %; or a total of Ti and Nb: 0.01-0.5 mass %.

Kindly replace paragraph [0035] of the Specification as published with the following:

~~In addition, in accordance with the above described aspect, the present invention~~ provides we provide a stainless steel for a proton-exchange membrane fuel cell separator wherein, of oxygens contained in the passive film, an atomic ratio of O(M)/O(H) between an oxygen O(M) present in the state of a metal oxide and an oxygen O(H) present in the state of a metal hydroxide is 0.9 or less.

Kindly replace paragraph [0036] of the Specification as published with the following:

~~Further, in accordance with the above described aspect, the present invention provides~~ we provide a stainless steel for a proton-exchange membrane fuel cell separator, wherein the Cr content is 20 to 45 mass %.

Kindly replace paragraph [0037] of the Specification as published with the following:

~~In another aspect, the present invention provides~~ We also provide a proton-exchange membrane fuel cell formed to comprise a solid polymer film, an electrode, and a separator,

wherein any one of the above-described stainless steels is used for the separator.

Kindly replace paragraph [0041] of the Specification as published with the following:

~~Best Mode for Carrying Out the Invention~~

Detailed Description

From the viewpoints of composition of stainless steels and composition of passive films, ~~the present inventor we~~ carried out extensive study and research regarding a stainless steel separator that would exhibit high corrosion resistance with contact resistance being restrained to be low. As a result, ~~the inventor we~~ discovered that with an Mo-containing high purity ferritic stainless steel being used as a base material, the composition of a passive film being generated on the surface thereof is adjusted, whereby the contact resistance is significantly reduced.

Kindly delete paragraph [0042] in the Specification as published:

~~To begin with, results of experimentation led to the present invention will be described herebelow.~~

Kindly replace paragraph [0043] of the Specification as published with the following:

As base materials, ~~the experimentation we~~ used cold rolled ferritic stainless steels (sheet thickness: 0.5 mm) containing 0.004 mass % C, 0.007 mass % N, 0.1 mass % Si, 0.1 mass % Mn, 30.5 mass % Cr, 1.85 mass % Mo, 0.03 mass % P, and 0.005 mass % S. Some of the base materials were subjected to annealing (950°C, 2 minutes) in the atmosphere and then to #600 wet polishing. Others of the base materials were subjected to annealing (950°C, 2 minutes) in a 75 vol. % of H₂ + 25 vol. % of N₂ ambient at a dewpoint of -60°C, whereby materials were prepared with a so-called bright annealing ("BA", hereafter) finish. Further, the base materials were subjected to etching at various temperatures for various time periods by using an acidic solution containing 10 mass % nitric acid, 50 mass % hydrochloric acid, and 1 mass % picric acid, and then to pure-water cleaning and cold-air drying for submittal to contact-resistance measurement. Concurrently, some of the wet-polished samples were subjected to pickling by being immersed in an acid mixture solution (8 mass % nitric acid and 2.5 mass % hydrofluoric acid, at 55°C) for 300 seconds, and are then subjected to pure-water cleaning and cold-air drying for submittal to the contact-resistance measurement.

Kindly replace paragraph [0053] of the Specification as published with the following:

To date, there have been cases of investigation conducted on the effects of the Cr/Fe atomic ratio of the passive film. However, knowledge regarding the mechanism having never been disclosed before has been obtained from the our experimentation. The knowledge teaches that adjusting the composition of the passive film enables the contact resistance to be significantly reduced. The mechanism is unclear, but can be envisaged as follows. The contact resistance is envisaged to be reduced for the reason that the increase in Cr/Fe atomic ratio increases the closeness of the passive film and reduces voids or voiding present in the passive film that interfere electrical conduction. In addition, the contact resistance is envisaged to be reduced due the reduction in O(M) / O(H) atomic ratio for the reason that the electrical conductivity of the hydroxide is relatively higher than the oxide. Further, the contact resistance is envisaged to be reduced due to the reduction in the Al content for the reason that the Al oxide having low electrical conductivity is reduced in the passive film.

Kindly replace paragraph [0070] of the Specification as published with the following:

~~In the present invention, in~~ In addition to the elements described above, Ca, Mg, REM (or, rear earth metal), and B may each be added in an amount of 0.1 mass % or less, or 1 mass % or less of Ni may be added to improve hot workability of the stainless steel being used as the base material of the separator. In addition, Ag in an amount of 1 mass % or less and Cu in an amount of 5 mass % or less may be added to decrease the contact resistance, and further, V may be added in an amount of 0.05 mass % to cause Ag to be finely dispersed.

Kindly replace paragraph [0072] of the Specification as published with the following:

Meanwhile, the steel ~~sheet~~ sheets are of the present invention is the ferritic stainless steel made of a ferrite structure composed of the above-described compositional ranges.

Kindly replace paragraph [0073] of the Specification as published with the following:

Properties and/or characteristics that the stainless steel for the separator ~~of the present~~ invention should own will now be described herebelow.

Kindly replace paragraph [0081] of the Specification as published with the following:

A preferred manufacturing method of the ~~inventive~~ our steel will now be described here. Conditions of manufacturing the ~~inventive~~ our steel are not specifically limited, but a general manufacturing method for ferritic stainless steels can be used. For example, preferably, respective steel is produced in such a manner that refining is conducted using a converter, and secondary refining is conducted in accordance with a strongly stirred vacuum-oxygen-decarburization (SS-VOD) process. As a casting process, a continuous casting process is preferable from the view points of productivity and quality. A respective slab produced by casing is heated to, for example, 1000-1250°C, and is hot rolled into a hot-rolled stainless sheet of a desired thickness. Preferably, after having undergoing hot-rolled-sheet annealing at 800-1150°C and hence pickling, the hot-rolled stainless sheet is further subjected to a step of cold rolling to be rolled to a predetermined product thickness; or alternatively, the cold-rolled sheet is further subjected to cold-rolled-sheet annealing at 800-1150°C or is further subjected to pickling, thereby being formed into the product steel. For the sake of productivity, two or more operations of cold rolling inclusive of inter-operation annealing may be performed by necessity in the above-described step of cold rolling. A total rolling-reduction rate in the step of cold rolling including one or two operations of cold rolling is set to 60% or higher, preferably 70% or higher. In addition, depending on the usage, low reduction temper rolling (such as skin pass rolling) moderate skin pass rolling is applied after cold-rolling and annealing. After a gas flow channel is formed by pressforming, the stainless steel sheet having thus been produced is preferably subjected to a process for adjustment of the passive film, thereby to be used as a separator.

Kindly replace paragraph [0084] of the Specification as published with the following:

~~According to the results of researches and investigations by the inventor, as shown in the conditions according to the present invention, for example, in~~ In the processing of a high Cr stainless steel by using a nitric acid+hydrochloric acid solution, the Cr/Fe atomic ratio was increased and O(M) / O(H) was decreased and favorable results could be gained when an acidic solution having a 2-10 times higher concentration of hydrochloric with respect to the concentration of nitric acid was used. In this case, it is preferable to perform the processing by

using the acidic solution to which a picric acid of 0.5-1.0 mass % is further added. Thereby, the effects of the processing can be gained in a short time.

Kindly replace paragraph [0085] of the Specification as published with the following:

In addition, in the case of a nitric acid+hydrofluoric acid solution, favorable results could be gained with an acidic solution having a 1.5 times higher concentration of the hydrofluoric acid with respect to the concentration of the nitric acid. In the case of each of the nitric acid+hydrochloric acid solution and the nitric acid+hydrofluoric acid solution, the preferred temperature of a acidic solution is 45°C or higher, and the processing time can be reduced to be shorter as the respective temperature is higher. However, processing liquids usable in the processing ~~according to the present invention~~ are not limited to those as exemplified above. In the event of the processing according to the immersion process, the condition may be selected in regard to, for example, the respective acid type, composition, temperature of the acidic solution and the processing time depending upon, for example, the composition and the surface finish of the base-material stainless steel. In the event of the processing according to the electrolytic process, the condition may be selected in regard to, for example, the composition of the respective electrolyte, electrolytic conditions, the temperature of the acidic solution, and the processing time.

Kindly replace paragraph [0103] of the Specification as published with the following:

Clearly in Table 4, also in the cases of respective BA-finished stainless steel sheets, unit cells using a separator for which the processing was performed using the liquid A or B on stainless steels satisfying ~~the our~~ compositional ranges ~~of the present invention~~ (specifically, Steel Nos. 3-6 and 9), and the Cr/Fe atomic ratio was set to 1 or greater and the Al/(Cr+Fe) atomic ratio was set to 0.10 or less by adjusting the composition of the passive film were each found to be as follows. The contact resistance is low, the initial output voltage, the output voltage after the passage of 2000 hours, equivalent to those of, for example, a gold-plated separator or a separator of a graphite plate can be gained. Thus, the respective unit cell can sufficiently withstand practical use.

Kindly replace paragraph [0105] of the Specification as published with the following:

However, in the case of any of stainless steels not satisfying ~~the our~~ compositional ranges ~~of the present invention~~ (specifically, Steel Nos. 1, 2, 7, and 8), the initial output voltage and the output voltage after the passage of 2000 hours are lower in comparison to the gold-plated separator or the separator of the graphite plate, regardless of the presence or absence of the composition adjustment process of the passive film.

Kindly replace paragraph [0106] of the Specification as published with the following:

Further, even in the case of each of the stainless steels satisfying ~~the our~~ compositional ranges ~~of the present invention~~ (specifically, Steel Nos. 3-6), when the composition adjustment process of the passive film is not performed or when the pickling generally used as pickling of the stainless steel is performed, the Cr/Fe atomic ratio of the passive film is low, and the initial output voltage is lower in comparison to the gold-plated separator or the separator of the graphite plate.

Kindly replace paragraph [0107] of the Specification as published with the following:

Industrial Applicability

~~According to the present invention, a~~ A stainless steel for a proton-exchange membrane fuel cell separator that has a low contact resistance and a high corrosion resistance can be obtained. For a proton-exchange membrane fuel cell which has used an expensive graphite separator because of durability problems, a low cost stainless steel separator can be provided.

Kindly replace paragraph [0108] of the Specification as published with the following:

~~The present invention~~ This disclosure is not limited to the proton-exchange membrane fuel cell separator, but can widely be used also as a stainless steel electrical component having electrical conductivity.